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Update on Monolithic Fuel Fabrication Methods

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ABSTRACT

Efforts to develop a viable monolithic research reactor fuel plate have continued at Idaho National Laboratory. These efforts have concentrated on both fabrication process refinement and scale-up to produce full sized fuel plates. Progress at INL has led to fabrication of hot isostatic pressed uranium-molybdenum bearing monolithic fuel plates. These miniplates are part of the RERTR-8 miniplate irradiation test. Further progress has also been made on friction stir weld processing which has been used to fabricate full size fuel plates which will be irradiated in the ATR and OSIRIS reactors.

Introduction

The United States Reduced Enrichment for Research and Test Reactors (RERTR) Fuel development program is tasked with the development of a fuel type that will allow conversion to low enriched uranium (LEU) of the world's research reactors that are currently fueled by uranium enriched to more than 20% U^{235} (HEU).

In order to accomplish this goal with little or no impact on reactor operation, the uranium loading of the new fuel must be raised to maintain a comparable fissile loading between the HEU and LEU fuel types. These efforts have led to metallic dispersion fuel (primarily uranium-molybdenum alloy with 7-12 wt.% Mo) and monolithic fuel where the fuel region consists of a single foil encased inside the aluminum cladding. While the U-Mo dispersion fuel represents an enhanced uranium loading (from a demonstrated 6 gU/cm^3 for U_3Si_2 to ~8 gU/cm^3) even greater loadings are needed for some high power reactors. These loadings can be increased by the use of the monolithic fuel which has a fuel meat loading of 16.3 gU/cm^3 for the U-7Mo alloy.

Unlike dispersion fuel where the fuel can be fabricated by the traditional roll bonding method, aluminum clad monolithic fuel must be formed by a different method[1]. Two methods are currently being pursued at Idaho National Laboratory (INL): friction stir welding (FSW) and hot isostatic pressing (HIP).

Hot Isostatic Pressing

-Experimental

Hot isostatic pressing uses simultaneous application of both heat and pressure to bond or densify materials. Implementation of the HIP process to clad U-Mo foils had been delayed by the lack of a HIP unit able to process uranium-bearing material (figure 1-left). Earlier out-of-pile tests at INL using a hot press to mimic the conditions found in the HIP were shown to achieve diffusion; this 'pseudo HIP' experiment showed interaction and bonding between the cladding and the fuel foil[2] as well as bonding between cladding materials.

A refurbished IPS HIP unit was installed in the RERTR fuel fabrication laboratory at the INL. This instrument has operating pressure and temperature limits of 207 MPa and 1100 ºC, respectively. It has a working zone 10 cm in diameter and 25 cm tall—large enough to fabricate RERTR miniplates (2.5 x 10 cm).

The fuel foil shape is identical to that used for FSW with a nominal thickness of 250 μm and an area measuring 1.9 cm x 8.25 cm with 0.32 cm radius corners. The cladding hardware is the same type used for the FSW process where a cover plate, slightly thicker than the target plate cladding thickness, sits on top of the foil which is placed in a recessed aluminum plate. The recess is cut to the dimensions of the fuel foil and is sized so the final cladding thickness will be the same on both sides of the foil. Unlike the FSW process which uses 6 x 15 cm plates (to provide room for clamping during assembly), the HIP plates are only slightly oversized at 5 x 15 cm. Prior to assembly, the plates are chemically cleaned in a process similar to dispersion fabrication[3]. The foils are cleaned by nitric acid.

The HIP process requires that the material be housed in a protective enclosure inside the pressure vessel to keep the pressing fluid media (argon gas) from interfering with the bonding surfaces. The HIP 'can' atmosphere is held at a partial vacuum to remove gasses that can hinder the bonding process. The enclosure or HIP 'can' is made of welded carbon steel and is designed to hold a total of six miniplates (figure 1-right). Tool steel strongbacks are placed on either sided of each plate to minimize process warping and the plates are isolated from the strongbacks and HIP can by layers of grafoil. The HIP can is built around the plate stack, after the plates are added the can is welded together. The integrity of the can is tested by helium leak check.

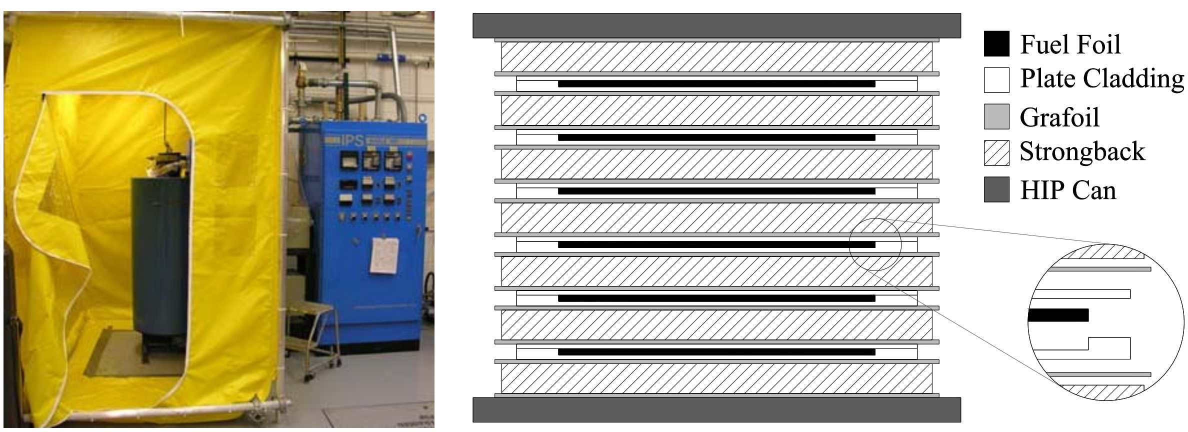

Figure 1. Miniplate HIP. Left: HIP unit with containment tent around pressure vessel (at left); Right: Cutaway schematic of the HIP can assembly.

The atmosphere is evacuated from the can by using a roughing pump to pull a vacuum on the can while it is heated. This bake out helps the bonding surfaces outgas and improves the surfaces of the bond. After a dwell time of 4 hours at 315 ºC the evacuation stem is crimped and welded.

-Processing

HIP process testing was conducted in four stages. First, aluminum cladding was tested to ensure that bonding could be achieved between the aluminum cladding material. Second, stainless steel foil surrogate blanks were used to test the ability to bond a stiffer foil and maintain cladding integrity and the required plate thickness and flatness. Third, depleted uranium alloy foils were included to examine interactions between the cladding and the fuel material. Finally the enriched fuel foils were included as feedstock for the irradiation experiment.

Aluminum 6061 alloy is used for all the RERTR irradiations and is the predominate alloy used in US designed research reactors. Testing on the aluminum cladding was done to find processing parameters that would achieve the required bonding with a minimal thermal signature to minimize the reaction layer formed between the fuel and the cladding. For the initial cladding tests a temperature of 580 ºC was chosen since it was just below the solidus temperature of 6061 (582 ºC). A heating rate of 290 ºC/hr and a cooling rate of 390 ºC/hr. were used. A pressure of 103 MPa was used during the hold period. The hold period was changed during the testing. Initially, a dwell time of 180 minutes was used. When this was shown to result in excessive interaction formation (below) this time was reduced to 90 minutes.

The HIP parameters were first tested on aluminum 6061 cladding and cladding bearing foils made of stainless steel surrogate. These tests determined that the HIP parameters were able to bond the aluminum cladding—which achieved a fully bonded fuel plate (figure 2-left). Surrogate tests with the stainless steel foil demonstrated that the HIP process is able to bond the aluminum cladding onto a foil of a harder material with a higher melting point (figure 2-right). Figure 2 shows the metallography of the surrogate plates. The interface regions of the aluminum/aluminum region show no remnants of a bond line. The microstructure is uniform across the entire thickness of the bonded aluminum. The quality of bond was not detrimentally impacted by the hold time reduction from 180 to 90 minutes. The stainless steel surrogate plate also shows full bonding in the aluminum/aluminum region. The foil/cladding interface shows some reaction layer as found in the previous 'pseudo HIP' run[2].

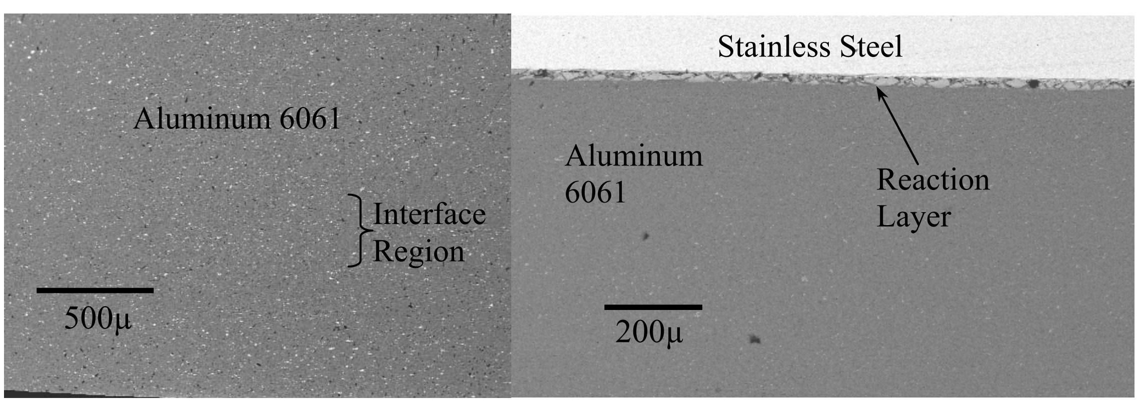

Figure 2. HIP bonded plates. Left: interface region of an aluminum/aluminum (both 6061) interface region. Right: stainless steel surrogate /aluminum plate with interaction layer. Both plates were processed at 580 ºC for 180 min.

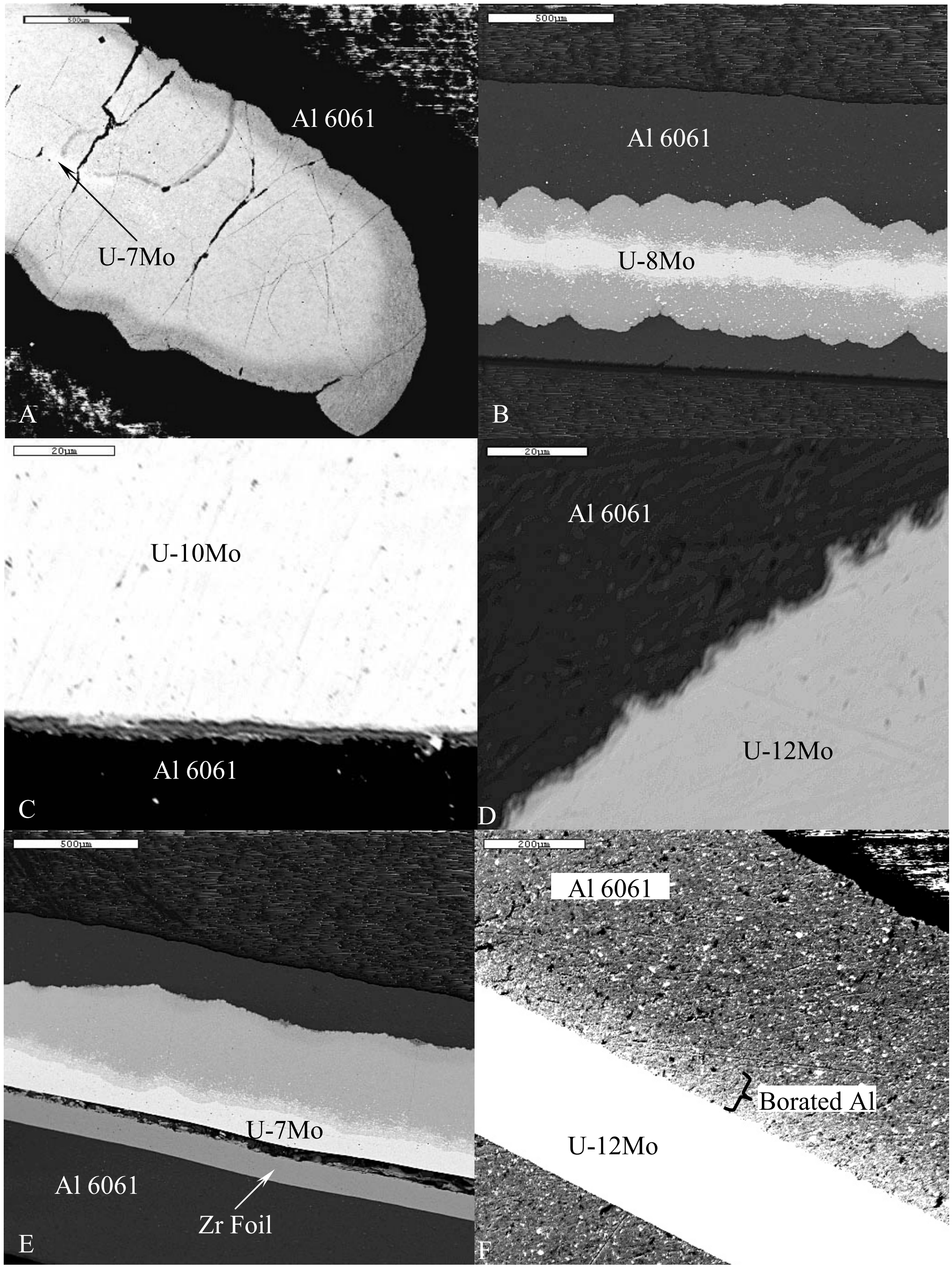

Figure 3. HIP process fuel plates. A: U-7Mo (180 min at HIP temperature); B: U-8Mo (90 min at temperature); C: U-10Mo (180 min); D: U-12Mo (90 min). HIP temperature for all plates was 580 ºC; E: U-7Mo fuel plate with Zr diffusion barrier (90 min); F: U-12Mo with layer of borated aluminum (90 min).

Figure 3 shows the micrographs of the HIP plates containing U-Mo foil. Initially, U-7Mo and U-10Mo foil compositions were tested with a HIP dwell time of 180 minutes. These plates showed a dramatic difference in interaction behavior. While the U-10Mo foil (figure 3-C) formed an interaction layer averaging less than 10 μm, the U-7Mo foil (figure 3-A) was virtually consumed. As these test plates were in the same HIP assembly, no experimental parameter excursion was deemed credible. Electron dispersive spectrography (EDS) of the U-7Mo plate shows the formation of two phase regions in the interaction layer: $(U\text{-}Mo)Al_3$ surrounding the small amount of the unreacted U-Mo alloy and a smaller band of $(U\text{-}Mo)_{0.9}Al_4$ adjacent to the remaining aluminum cladding.

The initial U-7Mo results showed that, if this fuel composition were to become a viable monolithic fuel, some way of minimizing the interaction layer must be found. One method is to reduce the severity of the thermal history (time at temperature). Further experiments were performed with a shorter dwell time (from 180 to 90 minutes). The composition range was also expanded to include U-8Mo and U-12 Mo foils. The U-8Mo foil tested at a 90 minute hold time showed extensive reaction but the center of the foil remained unreacted (Figure 3-B). The U-12Mo plate (tested in the same assembly as the U-8Mo foil) incurred only minimal interaction (figure 3-D).

The other method to limit the interaction between the fuel foil and the cladding material is to include a diffusion barrier at the foil/cladding interface to physically and chemically separate the two materials. In order to be successful, a diffusion barrier must be form a stable bond between both the cladding and the fuel material. Two materials were tried, zirconium and niobium. The material thicknesses used for these experiments were not of prototypic thickness—measuring 25 and 100 μm for the niobium and zirconium, respectively, but these materials did serve to examine the compatibility during fabrication.

UT Analysis of both of the fuel plates showed acceptable bonding through the thickness of the fuel plate. Micrographs showed a different story: The zirconium foil used as a diffusion barrier was bonded to the aluminum cladding but it also interacted with and separated from the U-Mo foil. This conflicting data is thought to be due to the formation of a brittle interlayer between the Zr and the U-Mo foil that maintained integrity during the UT scan but did not survive the cutting, mounting and polishing required for metallographic examination. Due to time constraints, the niobium bearing fuel plate has yet to be destructively examined.

Since some high power test reactors employ burnable poison to give a more favorable fuel performance lifetime, a fuel plate bearing a boron doped foil was included in the testing program. An aluminum 6351 alloy, impregnated with 1.04 weight percent boron, was rolled to a thickness of 75 μm. A section of this foil was cut to fit the recessed pocket in the monolithic hardware. The foil was placed between the fuel foil (U-12Mo) and the cladding and HIPed for 90 minutes. The micrograph (Figure 3-F) shows the two regions of aluminum next to the foil both of which show complete bonding. The magnification of the micrograph shown makes it impossible to make out the miniscule interaction between the U-12Mo and the cladding material.

Earlier results[4] have shown that the Mo content has a direct relationship with in-reactor stability. It has also been shown that the elevated Mo content alloys have a higher resistance to as-fabricated reaction layer formation[5]. This observation bears true for the HIP process where a clear difference is seen between the high (10 and 12 wt.% Mo) and the low (7 and 8 wt.% Mo) content alloys (table 1).

Table 1. Hot Isostatic Pressing of Uranium Plates

Fuel Alloy	Uranium Enrichment	Additional Material*	HIP Time (min)	Interaction Layer Thickness (μm)†
U-7Mo	DU	—	180	Foil fully consumed
U-7Mo	DU	Zr (100μm)	90	~400‡
U-7Mo	DU	Nb (25 μm)	90	Awaiting Examination
U-8Mo	DU	—	90	~250
U-10Mo	DU	—	180	<10
U-10Mo	EU	—	90	In RERTR-8
U-10Mo	DU	Al/B§ (75μm)	90	< 10‡
U-12Mo	DU	—	90	< 3
U-12Mo	EU	—	90	In RERTR-8
U-12Mo	DU	Al/B§ (75μm)	90	Awaiting Examination
U-12Mo	EU	Al/B§ (75μm)	90	In RERTR-8

* Placed adjacent to a single face of the U-Mo foil
† Average along face of foil where interaction layer exists
‡ Interaction layer thickness measured only on fuel/cladding interface
§ Borated aluminum (1.04 weight percent boron)

The initial study was far too short on resources (available U-Mo foils and time to conduct the experiments and still produce fuel plates for the RERTR-8 irradiation test) to conduct an exhaustive study. It is clear, however, that the molybdenum composition plays a definite role in reaction layer thickness irregardless of the length of the thermal histories tested. Direct comparisons between the two dwell times (90 and 180 minutes) can only be made with two examples. For both compositions the interaction data for the 90 minute run is taken from a fuel plate assembly where an additional material was included—the effect of this material on the measurements on the opposite side of the foil is judged to be, at most, trivial. Both U-7Mo and U-10Mo were examined after having undergone the two HIP histories. The U-10Mo foil reacted a total of less than 10 μm for both temperatures. The U-7Mo processed for 180 minutes was virtually fully consumed while the shorter run (which had only one face of the foil exposed) still had an average of less than half the U-7Mo unreacted.

-Bond Testing

Bond integrity was tested by three methods. First, was the non-destructive ultrasonic testing (UT). This was done to determine bond quality (debond testing) and the location of the foil inside the fuel plate ('minclad' testing). The second method is bend testing—where the cladding/cladding bond quality is examined by repeatedly bending it over a mandrel and examining for delamination. As this test is destructive in nature, it is performed on the adjacent material removed when the plate is cut to its final size. Finally, the bond can be examined by metallography. This examination looks for the presence of voids, brittle phases (which can be manifest upon sample preparation) and other anomalies.

The ultrasonic scans for all the HIP samples showed good bonding. Two representative sample are shown in figure 4. All of the HIP plates show surface effects caused by the grafoil being impinged irregularly into the surface of the aluminum during the bonding process. This rough ‘orange peel’ texture occurred on both faces of the plate and was removed only after 75-100 μm had been removed by machining the surfaces.

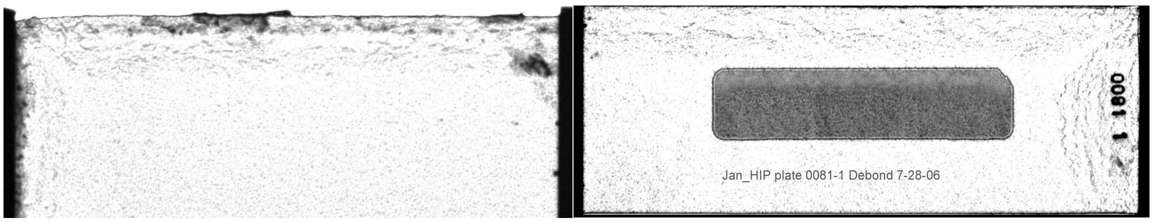

Figure 4. Ultrasonic debond scan. Left: UT scan of an unfueled aluminum cladding test. Right: UT scan of a U-12Mo fuel plate. The density variation between uranium and aluminum highlights the fuel foil. The mottled texture seen in both scans is due to the ‘orange peel’ surface finish, an artifact of the HIP process.

Bend testing for all the HIP plates was conducted on samples from around the perimeter of the fuel plate, in one position from each end and two samples from each side. The resulting samples showed no signs of delamination and were otherwise unremarkable.

The HIP process was used to produce three plates which are currently being irradiated in the RERTR-8 experiment in the ATR. Two additional plates containing lower molybdenum concentration alloys (7 and 8 wt.%) were originally planned to be fabricated by HIP but to avoid the heavily reacted plates, they were instead made by FSW.

Friction Stir Welding

The friction stir welding (FSW) technique remains largely unchanged since the last update on the process[6]. This fabrication method has been used to produce the majority of the irradiated monolithic miniplates. This method is also being used to fabricate the first full-size fuel plates for irradiation testing.

Two tests using prototypically-sized research reactor fuel plates are in the fabrication stages: IRIS-5 and AFIP. The IRIS-5 test is a joint venture between the French U-Mo group and the INL. Each will fabricate two monolithic fuel plates to be placed in the irradiation vehicle. The AFIP test, a joint venture between the INL and Argonne National Laboratory, is slated for testing in the ATR center flux position.

The plates fabricated at INL have shown good bonding across the fuel zone with only minor indications of less than full adhesion (figure 5-top). Regions of diminished sound transmission have been traced to flawed regions in the U-Mo foil itself (figure 6).

Minclad UT scans are taken to examine the interior of the fuel plate. The system generates a profile ‘slice’ every 25 μm through the thickness of the plate. These scans show foil bending inside the cladding. These foil ‘waves’ run along the length of the foil and correspond to the FSW tool passages over the plate (figure 5-bottom).

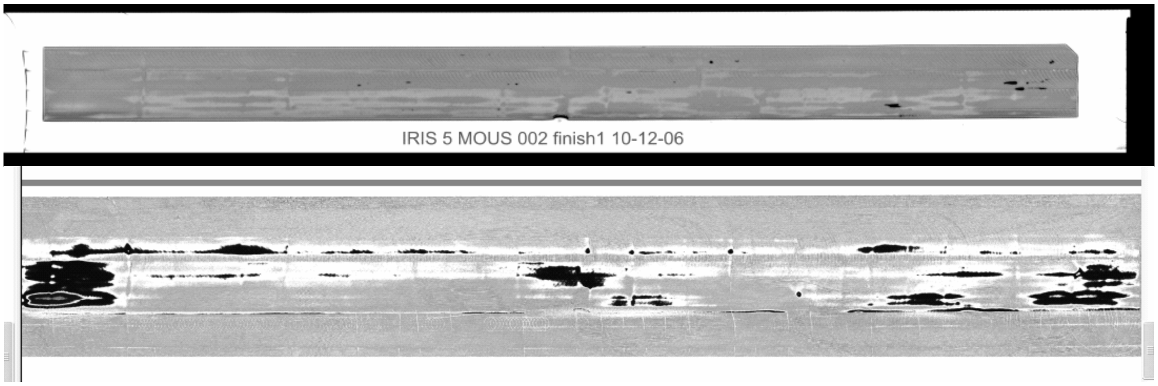

Figure 5. Ultrasonic scans of a full size monolithic fuel plate. Top: Debond scan. White regions are fully bonded aluminum cladding regions. The gray area is the fuel foil. Darker regions are regions of incomplete bonding; Bottom: Minclad scan. Scan shows a 'slice' through a thickness of the plate. Dark regions show the presence of the foil/cladding interface. Note the banding along the length of the plate indicating 'waves' in the foil caused by the FSW process.

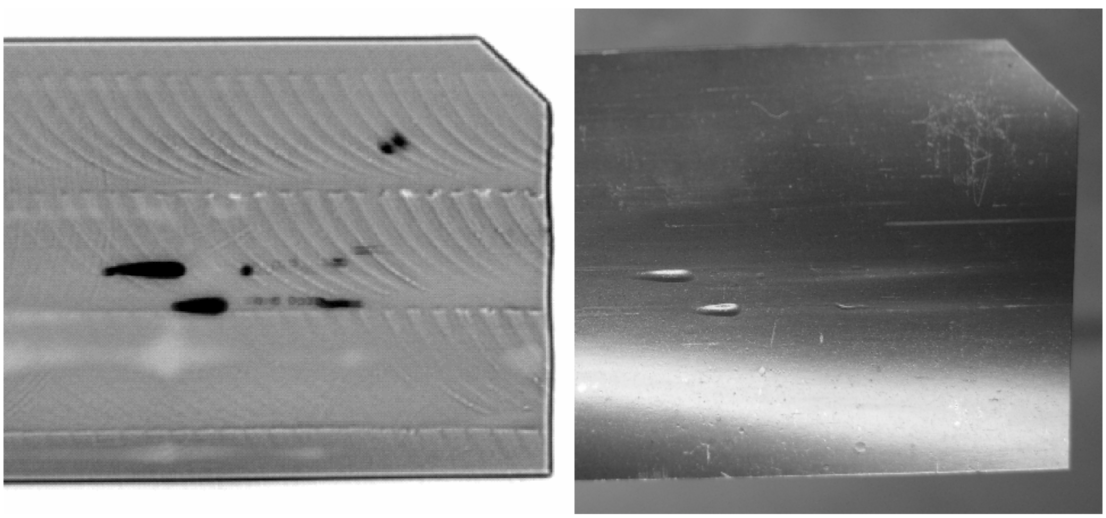

Figure 6. Detail of full-size monolithic fuel plate. Left: ultrasonic debond scan of part of the a LEU fuel plate fabricated for the IRIS-5 irradiation test. Black regions are areas of debond indications. Scalloped indications are surface features left by the FSW process. Right: photograph of foil prior to plate fabrication. Note the two blisters in the foil that correspond to the debond indication of the UT scan.

Transient Liquid Phase Bonding

The transient liquid phase bonding (TLPB) process was used to fabricate four miniplates irradiated in the RERTR-7a experiment. Of those four, two plates were included in the 'C' capsule which was found to have a fission product release. Visual in-canal examination (able to see only the one end of each plate and a foreshortened view of the plate faces) showed cracks on one end of each of the TLPB plates[7].

The plates in this experiment are scheduled to begin destructive examination in late 2006. No TLPB fuel plates were included in the RERTR-8 experiment.

Conclusions

The hot isostatic process as-fabricated results show great promise in monolithic fuel production. There is a clear difference in quality between the lower and higher molybdenum content compositions (between 8 and 10 wt.% Mo) with the HIP parameters tested. The higher Mo compositions show only minimal reaction while the lower reaction alloy compositions show an unacceptable amount of reaction layer formation.

Tests to limit the reaction layer by reducing the thermal history and including a diffusion barrier either did not show any significant thickness reduction (reduced thermal history), or did not achieve acceptable bonding between the barrier and the fuel foil. The reduced time at temperature, did provide adequately bonded plates, however and, after the initial testing, was used successfully on the remainder of the fuel plates.

The HIP process was used to fabricate three fuel plates that are being irradiated in the RERTR-8 test. Post irradiation test results are expected in 2007.

Additional work needs to be done on the HIP process to further study the interaction behavior and the effects of the process parameters. Work must also be done to scale-up the process to determine if it can be use to produce fuel plates on a commercially viable scale.

Friction stir welding is being used to fabricate the IRIS-5 and AFIP-1 full size monolithic fuel plate irradiation experiments. The plates for each experiment measure over one half meter in length and a width comparable to actual fuel plates. Initial results show that fuel plates can be made that satisfy reactor compliance criteria. Irradiation of these experiments is scheduled to commence within the coming months.

Acknowledgements

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